IOP Conference Series: Materials Science and Engineering

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To cite this article: P D Sutrisna *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **543** 012079

Microfiltration of Oily Waste Water: A Study of Flux Decline and Feed Types

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Abstract. In recent years, membrane separation techniques gain an increasing interest to be applied in many industrial separation processes, such as the separation of produced water in petroleum industry and the treatment of used oil and used cooking oil. Used oil and used cooking oil consist of suspended solids in addition to emulsion. Both ultrafiltration and microfiltration have been used to separate oil from water. The challenges of oil-in-water emulsion separation include the possibility of big oil drops deforming through the slot or pore of membrane thus reduce the oil rejection. In addition, concentration polarisation and fouling phenomena are considered as negative attributes for membrane performances. This research applied flat sheet microfiltration membranes to treat used oil and used cooking oil. The performances of membranes in terms of flux reductions were evaluated and analysed using filtration model. Experimental results showed the decline of permeate flux with time indicating the formation of polarisation layer on the surface of the membrane. In addition, complete blocking and deep bed filtration explained the flux behavior of used oil and used cooking oil filtration, respectively.

1. Introduction

The treatment of waste water plays an important role in chemical industries in terms of operating cost and environmental protection issue. The successful treatment of waste water will reduce the operating cost and also improve the environment quality around the industries. Examples of waste that is produced by chemical industry are oil-in-water (O/W) emulsion and water-in-oil (W/O) emulsion [1-5]. These kinds of emulsions cannot be discharged directly into the body of water because the emulsions can reduce the amount of dissolved oxygen. In addition to chemical industries, O/W and W/O emulsions can also be produced by local or household activities, such as kitchen frying. Hence, it is considered very important to treat the emulsions before they can be discharged into the body of water.

Until recently, several techniques, such as chemical demulsification, gravity or centrifugal settling, and electrostatic demulsification, have been utilised on the treatment of emulsion [1]. In addition to those techniques, membrane separation technique also gains an increasing interest to be applied to treat emulsion. Microfiltration and ultrafiltration are used more widely than other types of membranes to treat emulsions. During the treatment of emulsions using membrane filter, there emerges the possibility of the deformation of particles or drops through the pores of membranes. Such deformation causes the decrease in the quality of permeates thus reduce the efficiency of separation [1]. Several studies have

been conducted to utilize microfiltration and ultrafiltration on the treatment of emulsion waste. Hlavacek [6] analyzed the demulsification process through microfiltration membrane. It was found that the demulsification process could produce water with only 30 ppm of oil, which was much lower than the concentration of oil in feed emulsion. Other study suggested to employ slotted surface filters that can produce very high permeate flux at very low trans membrane pressure [7,8]. This type of membrane was specially design to treat emulsions and it can reduce the possibility of membrane pore plugging and fouling during the treatment of emulsion.
Despite of the successful utilisation of microfiltration and ultrafiltration on the treatment of emulsion on laboratory scale, there are still limited successes of the realisation of these techniques on industrial scale. It is considered very important to find suitable model from laboratory scale that can be employed to predict the behavior of membranes when the membranes are applied to treat real emulsion on industrial scale. Research on the investigation of flux behavior during O/W and W/O emulsions have been conducted recently using synthetic emulsion. The flux decline during microfiltration of O/W emulsion using porous glass tube membranes was investigated by Ohya et al [9]. The flux decline can be explained by two stages or models, i.e. blocking filtration and cake filtration. On another line of research, Tien and Ramarao [10] evaluated four filtration models, i.e. cake filtration, deep bed filtration complete blocking, and intermediate blocking. These models were first developed by Hermans and Bredèe in 1936 [11]. In the proposed models, there are four cases that might appear during the filtration process. The cases include cake filtration, deep bed filtration, complete blocking, and intermediate blocking as described in Figure 1.

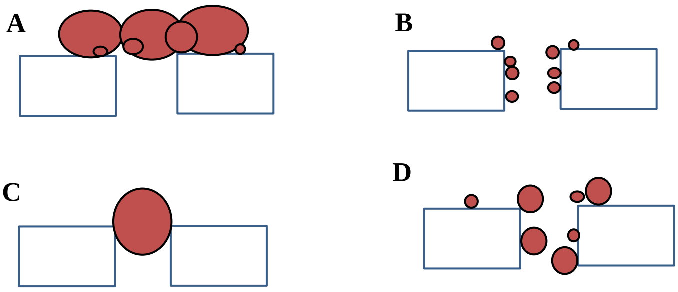

Figure 1. Schematic representation of filtration model: (A) cake filtration, (B) deep bed filtration, (C) complete blocking, and (D) intermediate blocking

In case A, the separation process is conducted with 40 wt% slurry that can form cake layer on the surface of filter. Case B explains the separation process using filter with relatively big pore sizes thus many particles will pass through to the permeate side. In case C, the filtration is done by using filter with small pore sizes, while case D using filter with pore sizes in between the pore sizes used in case 3 and case 1 or 2. All cases can also be explained by mathematical expressions as follows [10]:
Case A, cake filtration:
Filtration rate:

$$\frac{dV}{dt} = \frac{P}{\mu(AV+B)} \tag{1}$$

The correlation between V and t:

$$t = \frac{\mu}{P}\left(\frac{AV^2}{2} + BV\right) \tag{2}$$

Where V is volume of permeate, t is time, ⌠ is viscosity, and P is trans membrane pressure. A and B are constants.

Case B, deep bed filtration:
Filtration rate:

$$\frac{dV}{dt} = \left(\frac{dV}{dt}\right)_o (1 - aV)^2 = \frac{(dV/dt)_o t}{(1+a(dV/dt)_o t)^2} \tag{3}$$

The correlation between V and t:

$$V = \frac{(dV/dt)_o t}{(1+a(dV/dt)_o t)} \tag{4}$$

Case C, complete blocking:
Filtration rate:

$$\frac{dV}{dt} = \left(\frac{dV}{dt}\right)_o (1 - aV) \tag{5}$$

The correlation between V and t:

$$V = \frac{1}{a}\left[1 - exp\left\{-a\left(\frac{dV}{dt}\right)_o t\right\}\right] \tag{6}$$

Case D, intermediate blocking:
Filtration rate:

$$\frac{dV}{dt} = \frac{(dV/dt)_o}{(1+At)} \tag{7}$$

The correlation between V and t:

$$V = \frac{\left(\frac{dV}{dt}\right)_o}{A} ln(1 + At) \tag{8}$$

The correlations or equations (1-8) will be employed in this research to explain and predict the flux decline behavior during microfiltration of used oil and used cooking oil.

2. Materials and methods

Used oils in this research were collected from local workshop in Surabaya, Indonesia. Used cooking oils were gathered from local food stall in Surabaya, Indonesia. Before employed for filtration process, the viscosities of oils were analysed using Brookfield RVT. The viscosity of used oil was around 200 cP, while the viscosity of used cooking oil was around 125 cP. Whatman Cellulose nitrate microfiltration membranes were kindly supplied by Sigma Aldrich with nominal pore sizes around 0.2 ⌠m. The flat sheet membranes were soaked in aquadest before filtration process to remove some preservatives.
The filtration processes were conducted in a dead-end filtration set up that is presented in Figure 2.

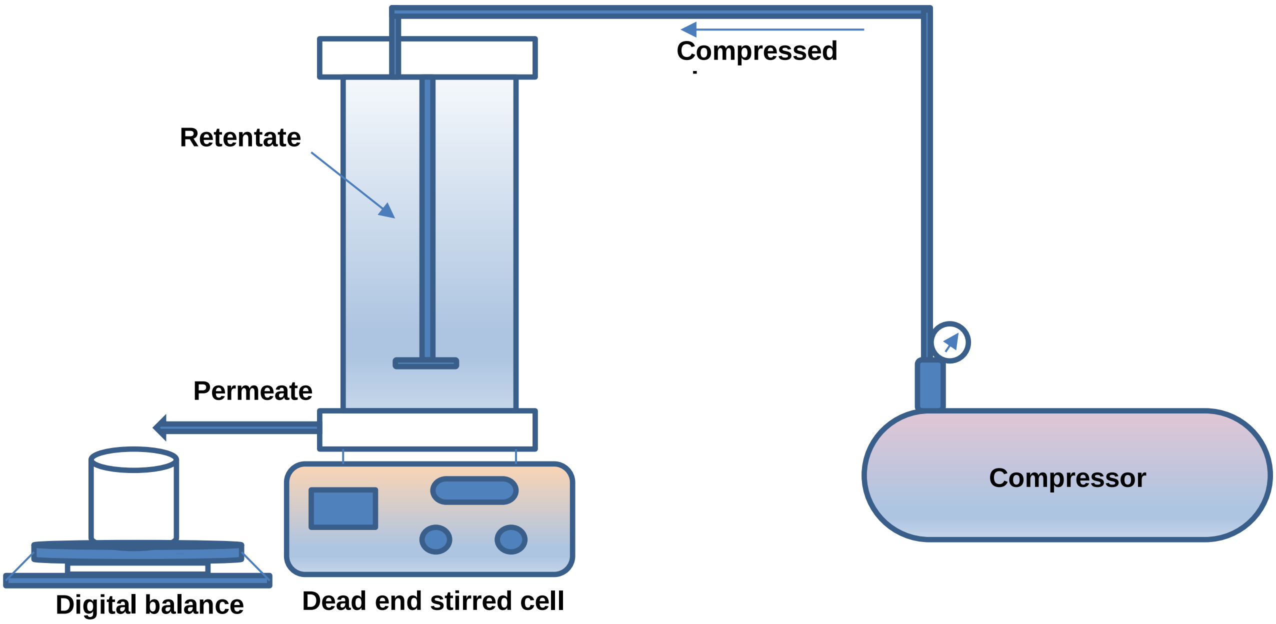

Figure 2. Experimental setup

The flat sheet membrane was placed inside the dead end stirred cell before it was contacted with feed emulsion. The emulsion was then poured into the cell and pressed with compressed air to induce feed pressure around 1 to 2 bar. During the filtration process, the permeate was collected inside a beaker glass and the flow rates of permeate were recorded. The flux of permeate through the membrane was then analysed and models based on equation (1-8) were generated. After each experiment, the membrane was dismantled and new membrane was then placed for the next experiment.

3. Results and discussions

3.1. *Flux – Time Profiles of Oil Emulsions Filtration*

In order to examine the influence of filtration time to the permeate flux, the relations of permeate flux and time for used oil and used cooking oil are presented in Figure 3.

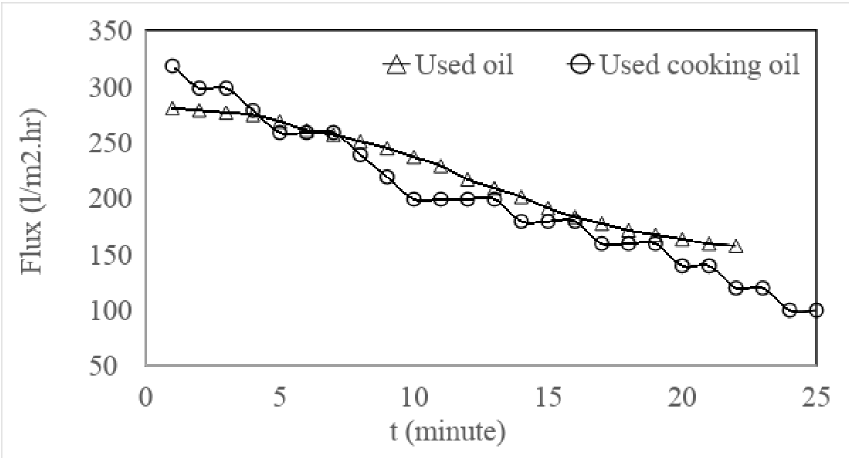

Figure 3. Permeate flux-time behavior during filtration of used oil and used cooking oil at feed pressure 1.2 bar.

The flux-time profile of used oil showed a relatively lower initial flux (~ 280 $l.m^{-2}.hr^{-1}$) compared to used cooking oil indicating the influence of solution viscosity and also the polarisation phenomenon. As mentioned in Section 2, the initial viscosity of used oil around 200 cP was higher than the viscosity of used cooking oil. This higher viscosity of used oil contributed to the lower movement of permeates through the membrane at the same trans membrane pressure. In terms of the rate of flux decline, the used cooking oil showed faster decrease in flux compared to used oil. This faster decline implies an accelerated polarisation concentration phenomenon during the filtration of used cooking oil. In short period after the experiment was conducted, there was non-blocking condition, and where the movement of the dispersed phase was only affected by the shear force generated on the surface of the membrane by the stirrer. Then, the flux behavior through the membranes will be determined by the formation of secondary membrane (dynamic membrane) on the surface of the membrane. This secondary membrane is always formed, whether automatically or by design from constituents in the feed. The formation of the secondary membrane will alter the performance of the primary membrane, in terms of flux and rejection. The formation of secondary membrane can be regarded as a beneficial effect, but this type of filtration behavior is usually associated with increasing filtration pressure, or decreasing filtration flux rates. Under these circumstances, the filtration is determined by the characteristics of the secondary membrane formed on the surface of the original membranes and, to some extent, the nature of the original membranes becomes less relevant.

Polarisation concentration will happen more severe if the feed solution contains more visible particles that can accumulate on the surface of the membranes. This phenomenon will hamper the utilisation of membrane on industrial scale because it will determine the life time of membranes. To extend the life time of membranes, polarisation concentration phenomenon needs to be handled and reduced. However,

IOP Conf. Series: Materials Science and Engineering **543** (2019) 012079 doi:10.1088/1757-899X/543/1/012079

to select the most appropriate technique to reduce the polarisation concentration on the surface of the membrane, we need to examine the mechanism of pore blocking and cake formation on the surface of the membrane. Compared to literature values, the permeate flux from this research showed comparable value as presented in Table 1. The concentration of oil used in this research was much higher than other studies indicating the potentiality of the membranes in this research for O/W emulsion separation.

Table 1. The comparison of permeate flux from several studies

Membrane material	Oil concentration (ppm)	Permeate flux ($l.m^{-2}.hr^{-1}$)	Ref.
Polyacrylonitrile (PAN)	1,000	100 – 2,000	(12)
2 microns true surface filter	1,000	200	(13)
7.5 microns slotted pore filter	1,000	1,820	(8)
0.2 microns cellulose nitrate	> 1,000	280 - 320	This work

3.2. *Mathematical Modelling of Flux Decline During Filtration*

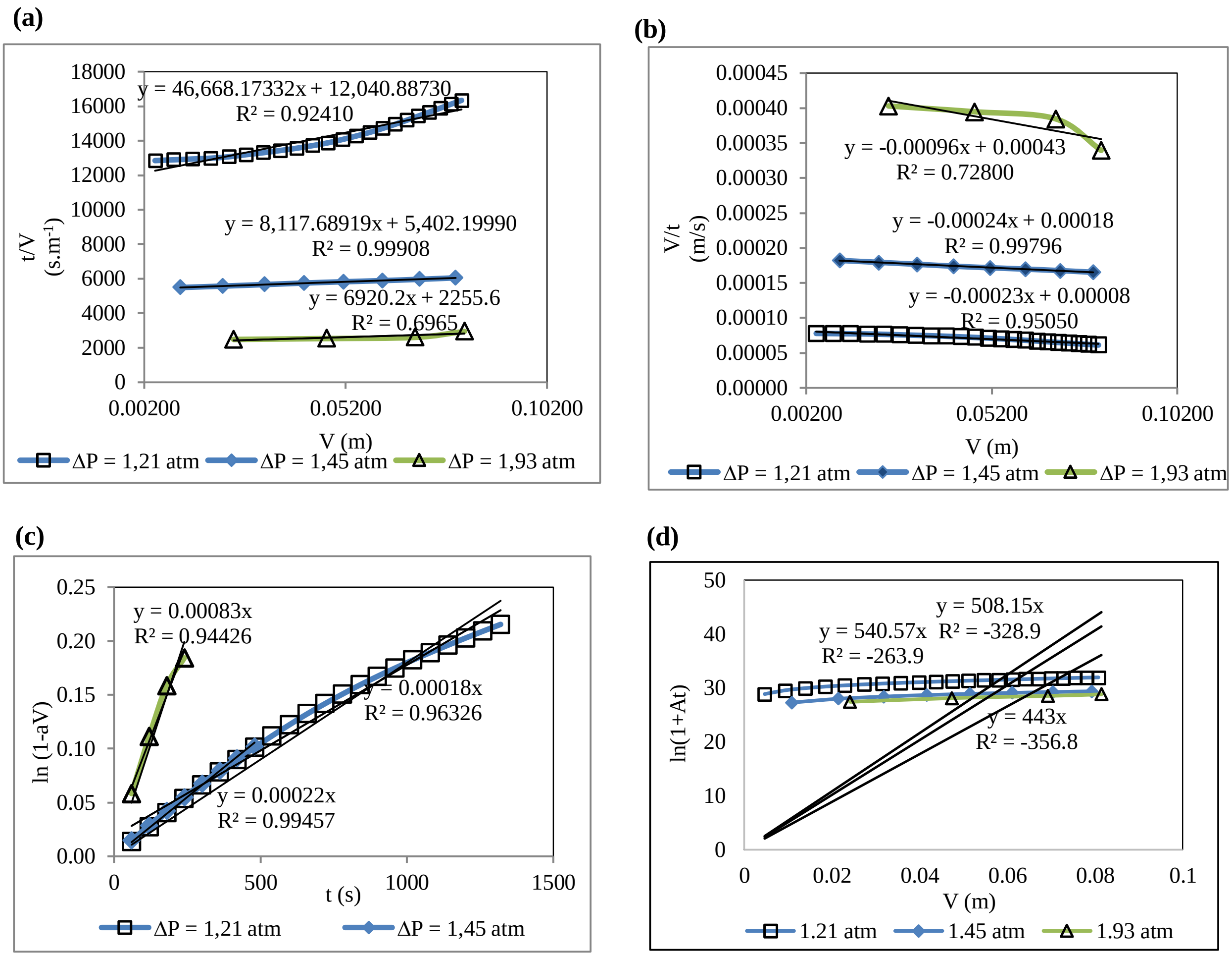

Figure 4. The results of mathematical modelling of (a) cake filtration, (b) deep bed filtration, (c) complete blocking, and (d) intermediate blocking during used oil filtration test

The correlations shown in Section 1 were used to explain the flux decline during the filtration of used oil and used cooking oil. Equation (2), (4), (6), and (8) were used and the results of the model for used oil are depicted in Figure 4.
According to the model, used oil filtration followed type 3 or complete blocking filtration model because the coefficient correlation (R^2) of that model showed the highest value at all feed pressure. Type 3 filtration model explains the blockage of membrane pores by large particles as shown in Figure 1C. Hence, the behavior of flux during used oil filtration was influenced by the presence of these large particles. The complete blocking of the pores by particles or dispersed phase could possibly decrease the flux through the membranes and the membrane can suffer from fouling phenomenon. In terms of feed pressure, the models suggest no influence of feed pressure on the flux behavior. As can be seen from Figure 4 the range of pressure used during the experiments need to be increased if one need to observe the influence of feed pressure on the flux behavior.
On the other hand, for used cooking oil, the flux behavior during experiment showed case 2 or deep bed filtration model as presented in Table 2. Deep bed filtration model explains the strong interaction between the wall of membrane pores and small particles, thus small particles will attach into the wall of membrane pores. However, this phenomenon also results in the decontamination of permeate by some small particles or dispersed phase.

Table 2. The results of modelling during used oil and used cooking oil microfiltration

Type of emulsion	⊗P (atm)	The type of filtration model	Correlation coefficient ,R^2
Used oil	1,21	3	0,9633
	1,45	3	0,9996
	1,93	3	0,9443
Used cooking oil	1,21	2	0,9966
	1,45	2	0,9902
	1,93	2	0,9638

4. Conclusions

The efforts to treat O/W and W/O emulsions play an important role to preserve the environment. Techniques such as membrane technology have been employed to separate the continuous and the dispersed phases of emulsions. Microfiltration with small pore size has been considered as a promising technique to treat oily waste water. However, its real application still needs to be explored and one particular concern is polarisation concentration phenomenon. In order to tackle this problem, the behavior of flux decline during filtration needs to be studied. This research was conducted to analysed and proposed filtration modeling of oily waste water using used oil and used cooking oil as feed emulsions. Experimental results suggested complete blocking and deep bed filtration behaviors for used oil and used cooking oil, respectively. The results of this research could bring an information to select suitable technique to reduce polarisation concentration in order to realise the implementation of the treatment of oily waste water by using microfiltration.

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